

CHAPTER 35

Use of Liquid Scintillation in the Appraisal of Non-Radioactive Waste Shipments from Nuclear Facilities*

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ABSTRACT

The generation of non-nuclear waste at the Savannah River Site (SRS) presents a special problem for analysis. As all waste must be suspected to contain radioactive materials prior to appraisal, large volumes of waste are retained on site prior to off-site shipment for incineration or burial. The amount of flammable waste solvent stored on site in late 1987 threatened to place the Savannah River Site in violation of EPA and OSHA regulations. One of the major contributors to this burden was some 2000 55-gallon drums of used paint solvent and waste paint. The Environmental Technology Section at the Savannah River Laboratory was charged with development of quick, reliable, and simple method for measurement of the maximum possible activity of paint waste samples. The <2 nCi/g guideline set by the Department of Transportation* is the upper limit of the radioisotope contamination for removal of the waste from the site.

Owing to the possible presence of tritium and the desire for a relatively quick analysis technique, liquid scintillation was used as the method of choice. Novel methods are presented for dealing with various phases (organic, aqueous, and solid) present in the samples and for determining the optimum dilution for severely quenched samples. Arguments are presented for the fragmentation of solid particles by ultrasonic vibration, and mathematical corrections are given for the escape fraction from solid particles.

INTRODUCTION

Nuclear facilities generate waste materials that must be removed from the site or disposed of on site. The characterization, isolation, and confinement of high level nuclear waste is currently the subject of much study. By contrast,

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*Code of Federal Regulations, 49CF173.403(g), June 1986.

relatively little attention has been given to non-nuclear waste from nuclear facilities.

On the Savannah River Site (SRS), nonradioactive or clean waste is normally assayed with a survey meter to appraise the extent of surface contamination. Because of the nature of the operations conducted at SRS, the majority of the waste generated is treated as contaminated waste until proven otherwise. Before such waste can be shipped off site, more rigorous assay must demonstrate compliance with government standards. In particular, it is necessary to appraise the samples with regard to the Department of Transportation (DOT) guideline for a total activity of 2 nCi/g of waste material.¹ Furthermore, the assay methods must be sufficient to comply with Environmental Protection Agency (EPA) regulations to prevent overcrowding of waste storage locations; thus, the present work developed efficient liquid scintillation methods for meeting these requirements.

BACKGROUND

The waste material at SRS is accumulated in 55-gallon drums and stored in interim storage facilities. These drums must be assayed in a relatively short period of time so that off-plant shipment can proceed in an orderly fashion, and the storage inventory complies with EPA regulations. All of this material was considered incinerateable nonradioactive hazardous waste. The present work examines 900 55-gallon drums containing paint solvent/waste which were to be shipped off plant for incineration. These drums contained up to three distinct phases of material with widely differing components. The materials were primarily mixed hydrocarbon solvents. A significant fraction of the material included paint solids, dust, and dirt. Additionally, many of the materials contained a measurable amount of water. All the samples contained the liquid organic material, but the other two phases were not always present. For these samples, neither the specific composition nor the contaminating isotopes could be assumed.

The radiometric methods used for these waste assays had to measure the total activity of waste samples in a relatively short turn around time. Sample preparation schemes such as fractional distillation for solvent cleanup, ashing of solid residue, digestion, solvent extraction, and either electroplating of a purified sample or pelletization of the original ash would have been simply too time consuming.

Methods that have been used for total low-level analysis of waste streams from other facilities also have such limitations. Traditional electroplating and alpha counting by semiconductor detector is a sufficiently sensitive method; however, this often requires lengthy chemical separations, and occasionally, sample ashing prior to plating of the sample. Gross plating of the original sample rarely proves satisfactory for such situations owing to the mixed character of the material and the time required. Because a large number of samples

must be processed on a continuing basis, this was not considered a viable alternative.

Tritium was considered one of the most likely contaminants of liquid waste materials resulting from on-site, processes. On-line methods for tritium detection in organic matrices do exist,² however, because much of the waste also contained a significant fraction of solid material, neither the on-line method previously mentioned nor simple liquid scintillation of untreated samples could be relied upon.

The total radioactive concentrations of the various waste materials had to be measured; thus, analysis for individual isotopes was not required. Based on the SRS process history, the major radionuclides were expected to be alpha, beta, and gamma emitters, although minor contributions from electron capture and positron decay could not be ruled out. Primary manmade radionuclides could be fission products, tritium, transuranics including plutonium, and neutron activation products. Liquid scintillation is very efficient in detecting the charged particle emitters and thus yields the total activity for samples devoid of radionuclides that decay by electron capture. Some of the neutron activation products do decay by electron capture, and their activities are best appraised by gamma spectroscopy due to their weak charged-particle emissions. The percentage of activity due to electron capture was judged to be fairly low and therefore liquid scintillation was enlisted as the primary technique for determination of "total activity." High purity, germanium diode, gamma spectrometry (HPGe spectrometry) was used as a consistency check to ensure that assumptions concerning ec-gamma activity were correct. HPGe spectrometry of the samples used routine methods; however, liquid scintillation required some method development to overcome the various problems associated with these samples.

Liquid scintillation can provide a measure of the total activity of a sample. All ionizing radiation can be detected by liquid scintillation; thus all isotopes could be expected to yield some degree of signal in a liquid scintillation sample. Of course, the detection limit varies for counting time and isotope of interest. As an example, tritium is one of the lowest energy beta emitters and its detection limit on a modern beta liquid scintillation spectrometer is about 11 picoCuries per sample given that no quenching is present and that the sample is colorless.

HPGe spectrometry is ideal for assay of gamma emitting nuclides, especially for gamma energies above 100 keV. The detection limits of HPGe spectrometry depend on the isotope and the counting geometry. In the Savannah River Site/Environmental Technology Section (SRS/ETS) Underground Counting Facility (UCF) the absolute sensitivity for ⁶⁰C in soil/water may be as low as 0.007 mBq/cm³, depending on sample configuration. This value is for a 24 hr counting time, and lower levels are attainable with longer counting periods. The samples must conform to a limited number of fixed geometries for the accurate use of HPGe spectrometry.

For many situations, low-level HPGe spectrometry is adequate for determi-

nation of total activity. In situations where the primary emission of the contaminate is a strong gamma-ray and the material can be made to conform to a known geometry, this technique is quite useful. Generally, little or no sample preparation is required for HPGe spectrometry. However, for appraising total activity, a significant amount of sample information is required prior to measurement. Also there must be a reasonable assurance that the primary contaminate is not tritium or some isotope such as ^{129}I which is not suitable for HPGe spectrometry.

ANALYSIS

Sample Preparation

Because of the large number of drums, it was decided to define an individual drum as a sampling unit and composite samples from these units were put in groups of $N = 2^n$ drums each. By using powers of two for the composite, a sample that was subdivided to trace a possible source of contamination (and minimize the amount of material that would have to be treated as low-level waste) could be subdivided down to a single barrel if necessary without the necessity of testing every single barrel in the original composite. This compositing allowed for much faster processing of the samples, but it did lead to increased background values as the dilution factor of N was now included in the detection limits. That made the acceptable maximum value for a single composite sample $2/N$ nCi/g. This smaller limit arose from the consideration that, in a single composite, there could be a single contaminated drum while all the other drums were clean. If N were too large, the analysis limit of $2/N$ would be too close to the detection limit of the instrument. Because of this, the maximum number of sampling units (55-gallon drums) in a single composite was determined by the background level of the instrument to be used. The number of sampling units in a single composite had to be selected so that $2/N$ nCi/g was higher than the background of the instrument. For most applications 16 or 20 was found to be a usable value for N . To create a composite sample, the drum contents were agitated, and then a long cylinder was used to retrieve a column of material extending from the top to the bottom of the drum. Uniform 25 mL aliquots were taken from each of these increments, and these subsamples were combined to give the analysis composite.

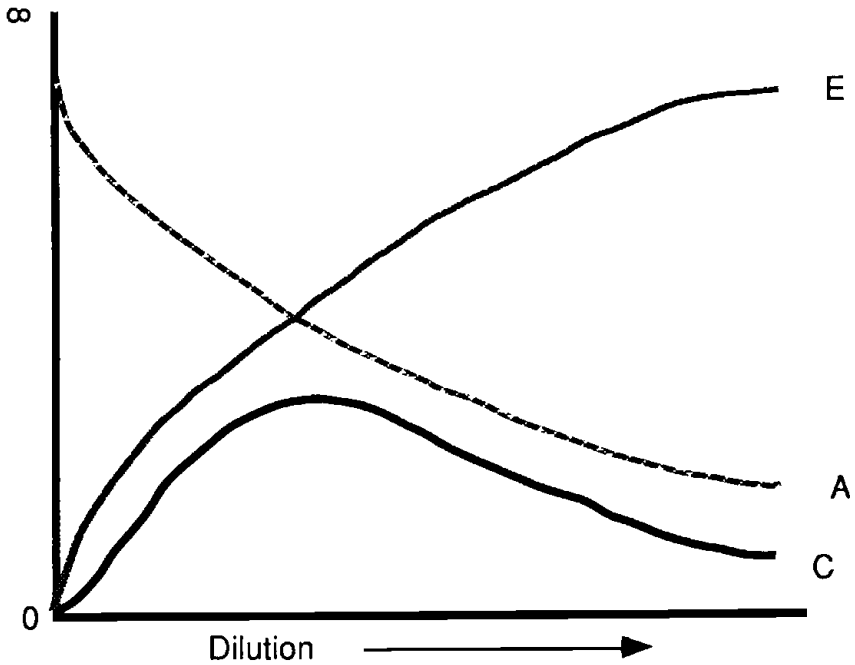
For the samples that consisted of multiple layers or phases of material, the components were separated into "homogeneous" samples for analysis; the entire sample could not be counted as a single entity or the heterogeneity would effect the results. After the phases were analyzed separately, the total activity would be found by factoring the relative masses of the phases into the summation of the individual activities. The relative masses of the phases were determined by measuring the volume (as a cylinder) of a single phase and using the empirically determined average density to calculate the mass of the phase in question.

For the organic liquid layer of the paint samples, quenching was dependent on the amount of the sample present in the cocktail. With the proper dilution of such a sample, it is possible to achieve an optimum count rate since both quenching and activity level would increase with concentration. Conceptually, this is shown in Figure 1. The resultant, optimum count rate, is the result of the increasing efficiency and the decreasing count rate with dilution. Assuming that the count rate (C), is the result of both the efficiency and the activity,

$$C = EA = Enm \tag{1}$$

where n is the specific activity and m is the mass.

$$\frac{\partial C}{\partial m} = En + \frac{\partial E}{\partial m}nm = 0 \tag{2}$$



E: Efficiency

A: Activity

C: Observed Count Rate

Figure 1. Graphic representation of dilution effects.

Substituting Equation 2 for the optimum mass we get,

$$m = m_0 = \frac{-E}{\partial E / \partial m} \quad (3)$$

To estimate an expression for E, it is assumed that the mass M of the sample in the cocktail reduces the scintillation light collection efficiency according to the Beer-Lambert law. A differential form of this law, as applied to the present work is

$$\frac{\partial E}{E} = -L \partial M \quad (4)$$

where L is a constant. This type of fractional decrease in efficiency with increasing sample mass is probably reasonable for a variety of quenching mechanisms; thus, Equation 4 was used for quenching in general as well as its integrated form

$$E = E_0^{-LM} \quad (5)$$

where E_0 is a constant. From Equations 3 and 4 it is seen that $M_0 = 1/L$. Thus M_0 could be determined by solving Equation 5 for L, using $E_0 = 1$ for the infinitely dilute ($M \sim 0$) case and $E = 0.001$ from tritium spike measurements. The resulting value for L is 2.24 gm^{-1} . Using this value for L a result of $\sim 0.4 \text{ g}$ was arrived at as the optimum sample size. With this as a starting point, a few empirical trials determined that the optimum dilution for the majority of the cases was $250 \mu\text{L}$ of organic material to a single cocktail. Each sample was combined with 20 mL of Opti-Fluor® scintillation cocktail and sufficient water to give a total volume of 23 mL.

Any aqueous phase was collected separately. This was achieved by reaching through the top organic layer with a disposable polyethylene pipette and transferring an aliquot of the aqueous material into a glass vial. The cocktail was then prepared out of this vial. The final sample preparation was identical to that for the organic layer with the exception that 1 to 1.5 mL of the sample was placed in the cocktail. In each case, the aqueous layer was less colored and exhibited less quenching than the organic layer.

Suspended solid materials presented the greatest problem. After examining several of the samples, it was discovered that most of the solids were chips of paint from 1 to 2 mm in "diameter." There were two primary concerns with this material: (1) most alpha events that occur in the chips themselves could be buried and never reach the scintillation cocktail, and (2) light resulting from alpha events in solution could be blocked or absorbed by the relatively large paint chips. Furthermore, the material was heterogeneous and not all particles were soluble in the Opti-Fluor® cocktail. Thus, it was necessary to reduce the size of the particles to the point where they would: (1) allow the majority of the alpha events to escape into the cocktail, (2) disperse homogeneously, and (3) remain suspended long enough to allow effective counting.

This dispersion and size reduction of the particles was accomplished by ultrasonically shattering the solids while they were immersed in a fairly viscous water/surfactant solution. For samples with a visible layer of solids, a polyethylene pipette was used to transfer 1.5 g of these solids to a tared glass vial. This material was diluted to a total volume of 10 mL with a 50/50 (v/v) mixture of Joy® detergent and deionized water. The vial was closed and immersed in an ultrasonic bath for a period of 15 min. One mL of this mixture was then used to prepare the liquid scintillation cocktail for evaluation of solids in the given sample. In previous applications at SRS laboratories, Joy® had been used for radiological cleanups and was found to harbor no abnormally high levels of activity. Also, blanks using tritium and Joy® showed no signs of quenching.

All samples were made up to a uniform volume of 23 mL. In all cases, both a normal sample and a spiked sample were prepared. For this work, the spike used was a dilution of a NBS standard tritiated water solution. A single spike consisted of 100 μ L of tritiated water having a disintegration rate of 32,000 dpm/mL. All samples were counted for three to nine periods of ten minute durations. The number of count periods depended on the time for any luminescence or chemiluminescence of the samples to decay. All samples were counted on a Packard Tri-CARB 2000 CA/LL with the output divided into three windows, 0 to 2 keV, 2 to 18 keV, and 18 to 2000 keV.

Sample preparation for the HPGe counting was much simpler. This preparation consisted of merely weighing the total sample and measuring the thicknesses of the individual layers in the bottle. In order to assure that any material on the outside of the bottles would not contaminate the SRS ultra low-level counting facilities, each bottle was placed in a tight-fitting polyethylene bag which was taped closed. The mass and height of material in each bottle was incorporated into the efficiency calculation for the HPGe detection.

ANALYSIS

Activity Calculations

As previously stated, the purpose of this work was to generate upper limit values which could be used as the basis for a decision for off-site shipment of hazardous waste. To this end, the liquid scintillation calculations were designed to yield the tritium and total radioactive concentrations. A tritium window of 2.0 to 18.6 keV and a total activity window of 0 to 2000 keV were selected for the basic analysis. Each sample and corresponding tritium-spiked sample yielded a tritium concentration (A_t), and a pseudo-total concentration (A_p). The value of A_p is larger than the actual total (A_a), because the tritium calibration spike is strongly quenched relative to other nuclides. Assuming that all other radionuclides are detected with an efficiency that is a factor of f greater than the tritium efficiency, the total activity concentration is given by:

$$A_a = A_t + \frac{(A_p - A_t)}{f} \quad (6)$$

In the present work, f is defined as the ratio of the ^{14}C and ^3H efficiencies³ as shown in Figure 2. Here the plotted quench parameter (tSIE) is defined for each sample from its spectrum with an external ^{133}Ba source.³ The magnitude of each efficiency curve is strongly correlated with the beta energy. Accordingly, the curve for ^3H betas, which range from 0 to 18 keV, is considerably lower than that for ^{14}C betas which range from 0 to 156 keV. Effectively, all other beta-emitting radionuclides have energies comparable to or greater than that for ^{14}C . Alpha particles are less efficient by a factor of 10 in converting their energies to scintillations; however, their higher energies (~ 5000 keV) result in scintillation responses that are well above those of the ^{14}C spectrum. Consequently, all radionuclides other than tritium are treated as having the same efficiency as ^{14}C ; thus making the overall calculation somewhat conservative.

The individual region of interest calculations of A_t and A_p used the following formula:

$$A = \left(\frac{S}{M} \right) * \left(\frac{C - C_0}{C_s - C} - \frac{B - B_0}{B_s - B} \right) \quad (7)$$

where $A = A_t$ or A_p for corresponding spectral window (dpm/g)

$S =$ Tritium spike activity (dpm/g)

$M =$ Mass of Sample (g)

$C/B =$ Sample/blank count rates

$C_0/B_0 =$ Sample/blank non-scintillant constant background rates

$C_s/B_s =$ Sample/blank tritium spike count rates

Calculation of Conservative Estimate and Best Estimate

For each composite sample, a conservative estimate (C_E) and a best estimate (B_E) were made for the total concentration of radioactivity in the entire sample. Both liquid and solid phases were treated and incorporated into a general formula:

$$E = F_l \times A_l + C_s \times F_s \times A_s \quad (8)$$

where $E =$ Either C_E or B_E , as appropriate

$F_l =$ Liquid fraction of sample

$A_l =$ Activity for liquid fraction (nCi/g)

$C_s =$ Solids correction factor

$F_s =$ Solids suspension fraction of sample

$A_s =$ Activity of solids suspension of sample (nCi/g)

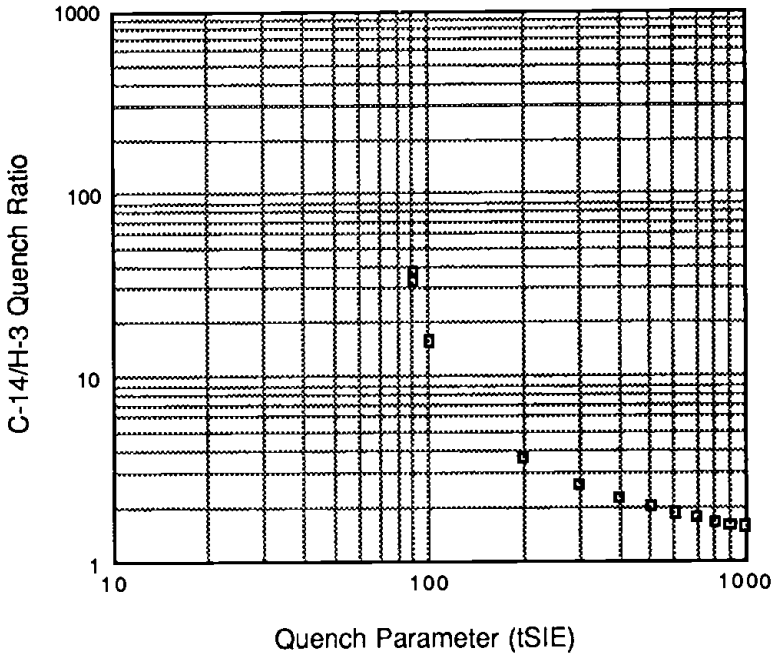
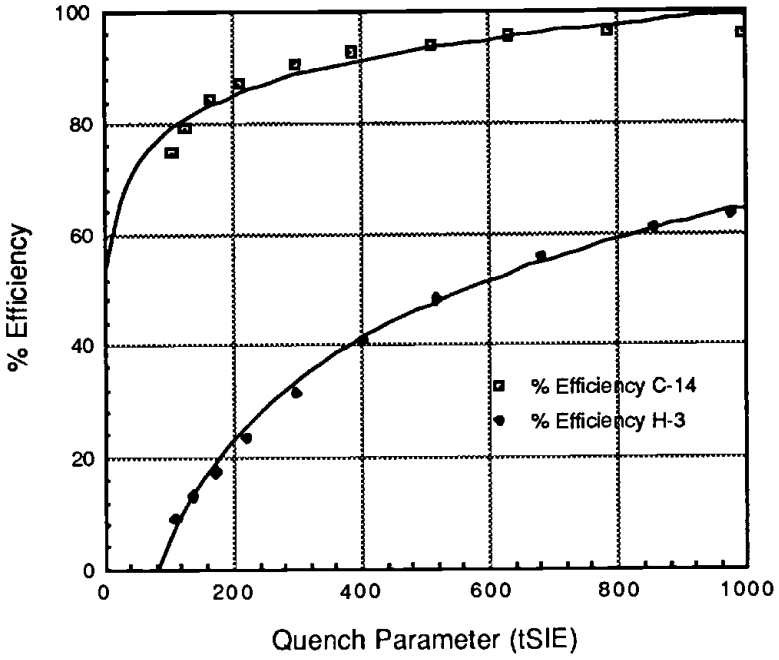


Figure 2. Individual isotope quench curves and ratio curve.

Here, only two phases are represented in the formula. In some cases, the formula was extended to handle an additional liquid phase (water).

The conservative estimate uses pseudo total A_p for activities A_1 and A_s , and a conservative correction (C_s) of four. As discussed earlier, A_p would be the total activity if all the count rate were assumed to be tritium. Because tritium is detected least efficiently, A_p is noticeably greater than the actual total activity A_a . The solids correction factor of 4 corresponds to a 25% escape probability for 4.5 MeV alphas uniformly distributed within a sphere of density 1 g/cm³ and a diameter of 200 μ M. Actually, the escape probability for tritium betas would be even lower, but tritium is not expected to be trapped in these solids after disintegration in the ultrasonic bath. Most other betas would escape more readily than the alphas. The correction of 4 is also conservatively high as the actual diameters of the particles are at or below 100 μ m, many alphas have higher energies than the values mentioned above, and no credit is taken for the distributional effects between solids and the suspending liquids.

The best estimate uses actual total A_a for activities A_1 and A_s , and a correction factor of $C_s = 1$. The use of A_a is an obvious selection; however, arguments for choosing $C_s = 1$ need to be reviewed. Here a 53% escape probability is calculated for 5 MeV alphas uniformly distributed within a 100 μ m sphere of density 1. However, any contamination for paint solids is likely to be distributed on the surface of the spheres, because the ultrasonic breakup of the paint solids should cause fractures along regions which have been exposed to potential contamination sources. If all activity is on the surface of the 100 μ m spheres, the escape probability increases to 69%. The solids suspension contains >60% liquid; thus, if the alpha activity were distributed proportionately to the solid and liquid fraction, the effective alpha escape probability would be >88%. The estimate should be even higher because (1) it is unlikely that all the activity is due to alphas (which have the shortest range in solids), (2) the typical liquid/solids ratio of >100 in the scintillation cocktail favors activity in the liquid, (3) many of the particles are likely to be smaller than 100 μ m in diameter, and (4) any internal solids activities are not likely to be from SRS sources and thus should contribute only a minor component from natural radiation. Adoption of $C_s = 1$ assumes that, taken together, each of the four additional factors contribute an average increase of only 3%.

RESULTS

The results of the individual samples are graphically shown in Figure 3 and listed numerically in Table 1. All values contain the average of at least three trials for liquid scintillation and the activity detected by HPGe analysis. Not only are the conservative and best estimates shown, but two additional values, the highest likely and highest possible activities, are shown. In these two additional cases, the assumption was made that all potential activity came from a single sample in the composite (highest possible), or that the original

Liquid Scintillation Results

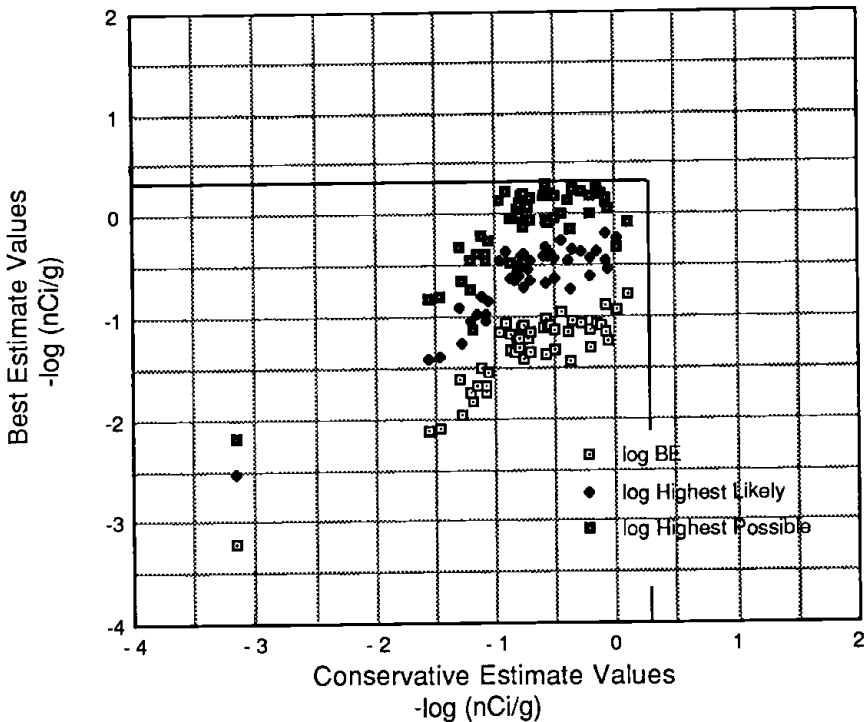


Figure 3. Plot of all composite and sub-composite samples (darker lines indicate limits for acceptance [\log of 2 nCi/g]).

distribution of possible activity in the samples was based on a log-normal distribution with 99% confidence (highest likely).

The highest possible estimate is the product of the best estimate activity and the number of samples in the composite:

$$\text{HPE} = N \times B_E \quad (9)$$

This represented the highest possible activity for one single drum in the composite. Only in the cases where both this value and the conservative estimate were both less than 2 nCi/g were the samples deemed acceptable for off-site release.

The highest likely estimate used a factor of 5.01 in the place of the number of samples, N . This value results from a log-normal probabilistic distribution of the material among the drums which make up the composite.⁵ These values are felt to be more realistic than the highest possible value above.

$$\text{HLE} = 5.01 \times B_E \quad (10)$$

Table 1. Numeric Results for all Composite and Sub-Composite Samples

Composite #	Samples	CE ^a	BE ^a	Highest Likely	Highest Possible
1	11	0.0007	0.0006	0.0030	0.0066
2	20	0.7600	0.0820	4.1080	1.6400
3	20	0.0280	0.0075	0.0376	0.1500
4	20	0.0510	0.0240	0.1202	0.4800
5	20	0.1070	0.0690	0.3457	1.3800
6	20	0.1250	0.0860	0.4309	1.7200
4	20	0.0340	0.0080	0.0401	0.1600
8	20	0.1580	0.0650	0.3257	1.3000
9	20	0.4370	0.0900	0.4509	1.8000
10	20	0.1520	0.0560	0.2806	1.1200
11	20	0.1340	0.0460	0.2305	0.9200
12	20	0.1590	0.0610	0.3056	1.2200
13	20	0.0620	0.0180	0.0902	0.3600
14	20	0.0830	0.0180	0.0902	0.3600
15	20	0.1660	0.0770	0.3858	1.5400
16	20	0.1750	0.0800	0.4008	1.6000
17	20	0.0860	0.0210	0.1052	0.4200
18	20	0.6230	0.0500	0.2505	1.0000
19	20	0.2930	0.0800	0.4008	1.6000
20	20	0.1750	0.0380	0.1904	0.7600
21	20	0.1880	0.0590	0.2956	1.1800
22	20	0.1960	0.0720	0.3607	1.4400
23	20	0.8430	0.0720	0.3607	1.4400
24	20	0.2010	0.0440	0.2204	0.8800
25	20	0.0870	0.0280	0.1403	0.5600
26	20	0.1440	0.0450	0.2254	0.9000
27	20	0.4160	0.0360	0.1804	0.7200
28	20	0.4160	0.0360	0.1804	0.7200
29	20	0.2650	0.0970	0.4860	1.9400
30	20	0.3180	0.0460	0.2305	0.9200
31	20	0.2620	0.0430	0.2154	0.8600
32A	10	0.0620	0.0180	0.0902	0.1800
32B	10	0.8160	0.1300	0.6513	1.3000
33	20	0.6930	0.0880	0.4409	1.7600
34	20	0.1600	0.0490	0.2455	0.9800
35A	10	0.2710	0.0820	0.4108	0.8200
35BA	5	0.1360	0.0670	0.3357	0.3350
35BB	5	1.2880	0.1670	0.8367	0.8350
36	20	0.3200	0.0750	0.3757	1.5000
37	20	0.0760	0.0310	0.1553	0.6200
38	20	0.2780	0.0770	0.3858	1.5400
39	20	0.6140	0.0750	0.3757	1.5000
40	9	0.3630	0.1110	0.5561	0.9990
41	20	0.8600	0.0570	0.2856	1.1400
42	20	0.2590	0.0760	0.3808	1.5200
43	20	0.5270	0.0840	0.4208	1.6800
44	20	0.4120	0.0700	0.3507	1.4000
45	20	0.0540	0.0110	0.0551	0.2200
46	4	1.0180	0.1180	0.5912	0.4720
47	20	0.0710	0.0210	0.1052	0.4200
48	5	0.0660	0.0150	0.0751	0.0750

^aCE and BE stand for conservation estimate and best estimate respectively.

The grounds for acceptance of a sample are illustrated by the diagram in Figure 3. If both the conservative estimate (CE) and the highest possible estimate (HPE) were below the 2 nCi/g guideline, the sample was accepted as below DOT guidelines. If the sample fell outside the DOT guidelines on either of the two above parameters, the samples in the composite were subdivided to create two new samples ($N_{\text{second}} = N/2$) and two new composites made up for analysis. This was required for two composites and one subcomposite. In fact, none of the samples which were outside the acceptance limits for the first composite were ever traced to a single "contaminated" drum. This fact supports the argument that the samples, which did initially fail to meet criteria, did so because of mathematical multiplication of background levels.

CONCLUSIONS

The method of phase separation, sample processing, and liquid scintillation analysis proved effective for the analysis of mixed phase, hydrocarbon based waste samples. The described method was readily applicable to a variety of circumstances and was straightforward enough to be used by staff analysts after a minimum of training. The results were obtained in far shorter time (usually less than 2 hr for a single sample) than would be required for other typical methods of analysis. All samples analyzed by the methods presented above were found to have activities below the 2 nCi/g DOT guideline for shipment of non-nuclear waste.

The effects of quenching and luminescence deserve further attention in the future. Methods for luminescence approximation through graphic interpretation have been considered, as well as the use of chemical agents to decrease luminescence effects. Overall, the compositing scheme reduced the number of samples almost by a factor of 20.

The ultrasonic fracturing of the solid particles provides a method for rapid analysis of this particularly difficult material. In the future, a more effective approach will be developed. Trial use of a cellular disintegration probe, as opposed to a ultrasonic bath, is underway at Waste Management Technology. Also, other materials such as Triton X-100 would probably provide better suspension characteristics than Joy.[®]

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